

INTERNAL STRESS AS A LINK BETWEEN MACROSCALE AND MESOSCALE MECHANICS

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1. Introduction

In this lecture note, the *internal stress* is defined as the stress that is maintained within a system by itself, without the aid of external supports or constraints.¹ From outside, the internal stress is, therefore, invisible by purely mechanical means. But since the stress exists locally on smaller scales, it could be observed, for example, by stress sensitive optical probes. Also, once the internal mechanical balance is somehow broken, the internal stress becomes visible from outside. In this sense, the internal stress is a concept bridging between macroscale and mesoscale mechanics. The purpose of this lecture is to draw the reader's attention to these aspects of the internal stress.

We can find the internal stress everywhere around us: When we tighten the belt of our clothes, the internal stress keeps contact between the belt under tension and our body under compression. When we make an espresso coffee, the internal stress is between the high-pressure vapor inside the coffee-machine and the stretched metallic frame of the machine. If we make a cut midway into a red sweet pepper or a watermelon, they generally change their shape, due to turgor pressure of plant cells which has accumulated the internal stress. When a pressing machine in industry spreads a metallic block, the framework of the machine and the block develop an internal stress, i.e., under compression in the metallic block on the one hand, under tension (and shear) in the machine's framework on the other hand. The *tensegrity* (Fuller, 1979) is a concept of self-standing architecture based on the internal stress. This is formed by wires under tension, rods under compression and the joints

¹ The internal stress in this context therefore includes the *residual stress* on the one hand, which has more strict definition than the former. Sometimes, however, the 'internal stress' is used just as opposed to the stress on the surface of a material (Landau and Lifshitz, 2002) on the other hand.



articulating those elements. The stable structure is realized by the internal stress among these them. This idea of tensegrity is applied to analyze the mechanical constitution in living cells (Ingber, 1997). The actin and micro-tubule filaments are thought to be joined by various active or passive binding proteins or other macromolecules. In solid clusters atoms interacts with each other by attractive and repulsive forces, making up a network of internal stress. Even the perfect salt crystal (NaCl) may be regarded as a result of internal stress due to electrostatic forces between like and unlike charge pairs. The atmosphere is pulled by the earth through the gravitaty, while the air also pushes the earth by the hydrostatic pressure. Any non-trivial self-sustaining system, therefore, contains the internal stress.

If the internal stress is found everywhere, is this concept a “general abstract nonsense” ?² Yes, perhaps. But this concept can provide with a useful viewpoint. I will try to develop this hope in the following sections: We first describe how the internal stress is maintained on the mesoscale (§2). Then we describe a mesoscale *sensor* that uses internal stress to realize the reliable observation under thermal fluctuations (§3). Next we discuss briefly the generation of internal stress in the context of far-from equilibrium process (§4). The last issue (§5) is the emergence of the internal stress as a memory effect.

I wish that this essay serves for the analysis, modeling and designing of various far from equilibrium phenomena such as glass dynamics, plasticity, and active media.

2. Mesoscale description of internal stress

2.1. INCOMPATIBLE STRESS-FREE STATES OF CONSTITUENT MODULES CAUSE THE INTERNAL STRESS

Under an internal stress, modules that constitute the system undergo some deformations. Let us look back some of the examples mentioned above. When a pressing machine presses down a metallic block, the latter is deformed from its natural form, and both the block and the machine develop the stress. Within a coffee maker, the water vapor is highly compressed with respect to the ambient pressure, and the container of the hot water/vapor opposes this pressure. The red sweet pepper before the cutting contains many cells whose shape is distorted with respect to their form under no constraints. Inside a cell many biopolymeric filaments are deformed by other components of the cell. In clusters or in crystals, many pairs of atoms are not at the distance of the lowest energy for a particular pair-interaction.

² The “general abstract nonsense” has been a word of criticism against the category theory (Mac Lane, 1998). This theory turned out to provide a powerful tool, as well as a conceptual perspective, to many field of mathematics.

Thus, on the mesoscale, the internal stress is caused by a quasi-static compromise among the constituent modules each of which insists on its own stress-free state³ (Dervaux and Amar, 2007).

2.2. INTERNAL STRESS IS THE CIRCULATION OF MOMENTUM FLUX

The state of a system having internal stress can be represented in terms of the momentum flux. We know that the total stress tensor σ with the minus sign carries the momentum. In (quasi-)static case of our interest the momentum flux density P , therefore, writes⁴

$$P = -\sigma. \quad (1)$$

The conservation law of momentum applied to the static state, $\nabla \cdot P = 0$, is nothing but the equation of static mechanical balance of the bulk without external force, $\nabla \cdot \sigma = 0$. If, for example, we focus on the flux of the x -component of momentum, $P \cdot \hat{x}$, the conservation law is $\nabla \cdot (P \cdot \hat{x}) = 0$, where \hat{x} is the unit vector along x -axis. This is mathematically of the same form as the conservation of mass flux, Fig. 1 (a) shows an example of the permanent momentum flux.

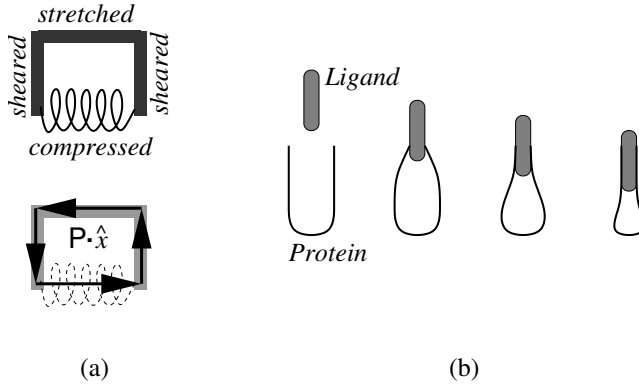


Figure 1. (a) (Top) An example of internal stress consisting of a spring and a framework. (Bottom) Schema of circulation of the flux of momentum in x direction, $P \cdot \hat{x}$. (b) “Induced fit” by *Protein* to a *Ligand*, realizing the arrival of signal to the conformational change of a receptor. Note that the ligand is under lateral stretching, while the bottom of the protein is under compression, as in (a).

³ If this static compromise becomes unstable, dynamical processes, such as an oscillation, explosion, chaos, can also occur.

⁴ In general case we should add the momentum flux associated with the material flow.

2.3. EXAMPLES IN SOFT MATERIALS

Surface buckling of swelling gels : Swelling of gel can take place by many different causes: it is sometimes enough to change the solvent (Tanaka et al., 1987) or the temperature (Doi, 2008). The swelling often develops a buckling pattern on the free surface. While the surface region of the gel can swell freely in the direction normal to the surface, it is laterally constrained in order to fit with the undeformed portion of the gel inside the bulk. The surface region of the gel is therefore under lateral compression. The buckling of the gel surface (Tanaka et al., 1987) occurs when the resulting lateral compressible stress exceeds a threshold (Sekimoto and Kawasaki, 1987; Suematsu et al., 1990). A similar phenomena occurs in the peripheral growth of membrane such as some vegetable leaf (Boudaoud, 2008). See also (Dervaux and Amar, 2007). Because of the freedom perpendicular to the membrane, the buckling pattern in this case occurs predominantly in this direction (Cerda et al., 2002). A complementary situation can also occur: If a gel is synthesized on a spherical substrate and pushed outward (Gerbal et al., 2000), the gel near the free outer surface develops a lateral tension, instead of compressive stress. This stress drives the lateral fracture of gel (Sekimoto et al., 2004; van der Gucht et al., 2005), instead of buckling.

Water suction at the crack tip of developing gel fracture : The velocity of gel's fracture is known to change if the solvent is supplied at the crack tip of developing gel fracture (Tanaka et al., 1996). The mode-I opening of the crack develops the negative pressure of the solvent within the gel. This negative pressure pulls the menisci of the solvent at the gel surface, which in turn compresses the gel toward inside. The internal stress establishes between the solvent under negative pressure and gel under compression. Recently (Baumberger et al., 2006) extrapolated the investigation of (Tanaka et al., 1996).

Drying gel : A similar situation occurs at the surface of drying gel or colloidal suspension. The surface layer of gel is under compression in the normal direction to the surface because it is pressed towards inside by the surface menisci of the solvent, on the one hand, and towards outside by the interior part of gel under osmotic pression⁵. A complementary situation oc-

⁵ The capillary pressure $p_{\text{cap}} = \gamma(K_1 + K_2)$, where γ is the surface tension and $\{K_1, K_2\}$ are the principal curvatures of the meniscus, creates the permeation flow obeying the Darcy's law, $\nabla p = -\zeta(1 - \phi_{\text{gel}})(\mathbf{u}_{\text{solv}} - \mathbf{v}_{\text{gel}})$, where ζ is a constant, ϕ_{gel} is the gel's volume fraction, and $\{\mathbf{u}_{\text{solv}}, \mathbf{v}_{\text{gel}}\}$ are the velocity of solvent and gel, respectively. The gel is pressed toward inside by the boundary condition, $\pi_{\text{gel}} = p_{\text{cap}}$, while it is pressed toward outside by the total mechanical balance in the gel, $\nabla \pi_{\text{gel}} = -\nabla p$, where π_{gel} is the osmotic pressure in the gel.

curs in ionic gels: The counterions between the network of polyelectrolyte gel (Khokhlov, 2008; Doi, 2008) push outward the free surface of the gel through the electrostatic double layer on the surface (Donnan effect) (Ricka and Tanaka, 1984), while the network elasticity resists against the swelling.

Buckling of drying colloidal suspension? On continuing the discussion of the drying gel, the gel layer under compression perpendicular to the surface wants to expand laterally along the surface. *If* this expansion is geometrically inhibited, there develops the compressive stress along the surface. ⁶ I suppose that this secondary effect is the origin of the surface pattern observed before the drying fracture (Ma et al., 2004), see Fig. 2.

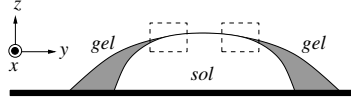


Figure 2. Schematic sectional view of a drying droplet of colloidal suspension. The shaded region is in the gel phase, while the central part of the droplet is still in the sol phase. In the regions marked by dashed rectangles, the compressed gel in z -direction develops also compressive stress in x -direction, while there is no compression along y -direction due to the sol-air interface in the central part of the droplet.

Permanent set of rubber crosslinking : Suppose we have a rubber network deformed by an external force. If we introduce further crosslinking into this deformed network, the network has virtually two networks whose stress free states are mutually incompatible. After removing the external force, the rubber returns to a new apparent “stress-free state.” This state is called the *permanent set* (Flory, 1960).⁷

We thus have seen that whether or not the internal stress is visible depends on the scale of description / observation.

⁶ Given the normal compressive stress, $-\sigma_{zz} > 0$, the induced lateral compressive stress is $-\sigma_{xx} = -\sigma_{yy} = -\frac{\nu}{1-\nu}\sigma_{zz}$ for isotropic surface, or $-\sigma_{xx} = -\nu - \sigma_{zz}$ and $-\sigma_{yy} = 0$ when the y direction is not constrained (Ma et al., 2004), where ν is the Poisson ratio of the gel’s osmotic elasticity.

⁷ The purely entropic free energy of original rubber writes $-TS_1 = (k_B T \nu_0 / 2)(\lambda_x^2 + \lambda_y^2 + \lambda_z^2)$, where λ_x etc. are the elongation ratios along x -direction etc. with $\lambda_x \lambda_y \lambda_z = 1$. Then, the free energy, $-T(S_1 + S_2)$, after the second crosslinking introduced under deformation, λ_{x0} etc. with $\lambda_{x0} \lambda_{y0} \lambda_{z0} = 1$ is such that $-TS_2 = (k_B T \nu_1 / 2)[(\lambda_x / \lambda_{x0})^2 + (\lambda_y / \lambda_{y0})^2 + (\lambda_z / \lambda_{z0})^2]$. Then the permanent set has the deformation, λ_{xs} etc. obeys $\nu_0 \lambda_{xs}^2 + \nu_1 (\lambda_{xs} / \lambda_{x0})^2 = C$ etc, where C is determined by the condition $\lambda_{xs} \lambda_{ys} \lambda_{zs} = 1$ (Berry et al., 1956). Macroscopically, the new rubber has free-energy, $-TS_s = (k_B T \nu_c / 2)[(\lambda_x / \lambda_{xs})^2 + (\lambda_y / \lambda_{ys})^2 + (\lambda_z / \lambda_{zs})^2]$, with $\nu_c^3 = (\nu_0 + \nu_1 / \lambda_{x0}^2)(\nu_0 + \nu_1 / \lambda_{y0}^2)(\nu_0 + \nu_1 / \lambda_{z0}^2)$. See also (Kaang et al., 1997).

3. Sensor working on the thermally fluctuating scale

The sensor of signal molecules (ligands) must fulfill apparently incompatible requirements: it must respond strongly enough to the arrival of the signal through the conformational change of the sensor itself, on the one hand, but the interaction between the ligand and the sensor must be mostly neutral to reflect the density of the ligands in the environment, on the other hand. The biological receptors solved this task using the mechanism called the *induced fit* (Koshland, 1973), which uses the internal stress (Sekimoto, 2005). Fig. 1(b) explains how the attraction or tension between the ligand and the sensor deforms the latter. The analogy to the Fig. 1(a) is evident. It is this deformation that represents the recognition of signal, satisfying the first requirement mentioned above. In energetic term, the gain of energy by binding interaction can be compensated by the cost of deformation of the sensor. This cancellation enables the bias-free arrival and departure of the ligand, satisfying the second requirement of the sensor.

We note that the reversible detection of signal described above has no contradiction with the theorem of Landauer and Bennett (Landauer, 1996; Bennett, 1982), telling that we need at least $k_B T \log 2$ to know the content of a single-bit memory. The latter process decreases the entropy by $\log 2$ of the “movable bit”, or, the observer at the compensating external work of $k_B T \log 2$. In the reversible detection, the system of ligand and detector is isolated from the external observer who gains no information.

4. Generation of the internal stress

In many cases, the states with internal stress are metastable. (The ionic crystal may be an exception.) To create these metastable states, we require far from equilibrium operations: The pressing machine turns a motor to raise the oil/air pressure in the piston. The coffee machine generates inner pressure by heating and boiling. Plant cells develop the pressure of vacuoles by pumps. Colloidal gel generates the stress through evaporation flux. The permanent set of rubber network is made by chemical reaction.

The generation of internal state as metastable state is closely related to the notion of the *plasticity*. It is because, during the generating process, some timescales of the system is momentarily diminished at the same time that the bias is put in favor of the (future) metastable state.⁸

In order to create the internal scale of a certain spatial scale, we do not necessarily need the operations on that scale. Suppose that we bend strongly

⁸ We may even compare with the positive discriminations to change the society’s stable state, because excessive forcing is required momentarily in both case.

a metal rod. The inhomogeneity and anisotropy on the atomistic scale inside the bar can generate the metastable states of small scale, such as entangled dislocation network, a typical example of internal stress. After removing the external forcing, the rod once yielded will show the different elasticity as well as the different yield stress (Ooshida and Sekimoto, 2005), as compared with those of the original state. The new metastable state reflects the preceding far from equilibrium operation.

5. Macroscale emergence of internal stress

We discussed above how an external macroscale forcing is “internalized” as the internal stress of mesoscale. Below we describe, to some detail, the opposite case: The internal state of mesoscale emerges as stress of macroscale through far from equilibrium processes.

5.1. RHEOLOGICAL MODEL OF RUBBER :

We take a simple phenomenological constitutive equation for a linear segment of rubber:

$$\sigma_t = G_R \gamma_t + G_\infty \int_0^t \mathcal{G}(t-t') \dot{\gamma}_{t'} dt', \quad (2)$$

where the tensile stress σ_t at time t consists of two terms. The first is a purely entropy-elastic stress, $G_R \gamma_t$, with γ_t being the elongation ($\lambda - 1$) at the same time and G_R being the rubber elastic modulus. The second represents the rheological response of network chains, where $\mathcal{G}(\dagger)$ is the relaxation kernel satisfying $\mathcal{G}(0) = 1$ and $\mathcal{G}(\infty) = 0$, and G_∞ is the lass elastic modulus. Mathematical structure of (2) is essentially the same as the constitutive equations of magnetic or dielectric materials. If $\mathcal{G}(z) = e^{-z}$, the model reduces to the well-known Maxwell model of rheology. We assume that $\gamma_t = 0$ for $t < 0$.

The following rewriting of (2) is physically appearing:

$$\sigma_t = G_R \gamma_t + G_\infty \left[\int_0^t \frac{\partial \mathcal{G}(t-t')}{\partial t'} (\gamma_t - \gamma_{t'}) dt' + \gamma_t \mathcal{G}(t) \right]. \quad (3)$$

If we interpret $G_\infty(\gamma_t - \gamma_{t'})$ as the force due to a Hooke spring created at the time t' , the multiplicative factor, $\frac{\partial \mathcal{G}(t-t')}{\partial t'} dt'$, is its survival probability until the present time t . The rheological response of network chains can, therefore, be viewed as the permanent set which is incessantly crosslinked but also uncrosslinked with the surviving probability $\mathcal{G}(\cdot)z$ until z sec after the crosslinking.

5.2. INTERNAL STRESS IN THE RHEOLOGICAL MODEL

Next suppose that we stopped stretching at time t_0 and let loose the sample. Shortly after releasing the stress ($t = t_0^+ \equiv t_0 + \epsilon$ with small positive ϵ), the stretching $\gamma_{t_0^+}$ ($0 < \gamma_{t_0^+} < \gamma_{t_0}$) should obey the following equation:

$$0 = G_R \gamma_{t_0^+} + G_\infty \left[\int_0^{t_0^+} \mathcal{G}(t_0^+ - t) \dot{\gamma}_{t'} dt' + (\gamma_{t_0^+} - \gamma_{t_0}) \right]. \quad (4)$$

Since the crosslinks have finite lifetimes, the vanishing of the left hand side of (4) is the result of internal stress established as compromise among the temporary crosslinks or Hooke springs. Some springs are under tension but some others should be under compression. This internal stress can remain for very long time if the temperature is below the glass temperature.

5.3. MEMORY EFFECT OF RUBBER

Suppose we then fix again the sample's at the relaxed length $\gamma_{t_0^+}$. Initially $\sigma_{t_0^+} = 0$ by definition. But what will be the stress σ_t for $t > t_0^+$? We can show that σ_t for $t > t_0^+$ under fixed $\gamma_t = \gamma_{t_0^+}$ writes as follows:

$$\begin{aligned} \sigma_t &= G_R \gamma_t + G_\infty \left[\int_0^{t_0} \mathcal{G}(t - t') \dot{\gamma}_{t'} dt' + \mathcal{G}(t - t_0) (\gamma_{t_0^+} - \gamma_{t_0}) \right] \\ &= G_R [1 - \mathcal{G}(t - t_0)] \gamma_{t_0^+} + G_\infty \int_0^{t_0} [\mathcal{G}(t - t') - \mathcal{G}(t - t_0) \mathcal{G}(t_0 - t')] \dot{\gamma}_{t'} dt' \end{aligned} \quad (5)$$

where we substituted $\mathcal{G}(t - t_0) \times (4)$ to go from the first line to the second. We note that $\mathcal{G}(t - t') - \mathcal{G}(t - t_0) \mathcal{G}(t_0 - t')$ in the last integral vanishes if $\mathcal{G}(z)$ is the Maxwell model, $\mathcal{G}(z) = e^{-z}$. Otherwise, a stress reappears before it returns finally to the rubber elasticity. This phenomenon was first found experimentally (Miyamoto et al., 2002). We call this reappearance of stress the memory effect. The Maxwell model, therefore, cannot explain the memory effect. Intuitively, if there is more than one characteristic times in the relaxation kernel $\mathcal{G}(z)$, the balance of internal stress on the mesoscale is transiently broken. And the uncompensated mesoscale stress appears as the macroscale stress. Due to the mathematical similarity of the constitutive equations, the memory effect in magnetic or dielectric systems is also understood with pertinent reinterpretation of the stress.

From information point of view, the *history* of operations in $0 < t < t_0$ can be read out from the memory effect in $t_0 < t$, up to the memory capacity

(n) of the system, where n is the number of characteristic times in $\mathcal{G}(z)$, i.e. $\mathcal{G}(z) = \sum_{j=1}^n a_j e^{-z/\tau_j}$.

From energetics point of view, we can assess the energy stocked in the state of internal stress. We use an expression of σ_t similar to (3), which is of course equivalent to (5):

$$\sigma_t = G_R \gamma_{t_0^+} + G_\infty \left[\int_0^{t_0} \frac{\partial \mathcal{G}(t_0 - t')}{\partial t'} [\gamma_{t_0^+} - \gamma_{t'}] dt' + \mathcal{G}(t) \gamma_{t_0^+} \right]. \quad (6)$$

By an analogy to the potential energy of Hooke spring, the internal energy E_t^{int} contained by the mesoscale springs should be

$$E_t^{\text{int}} \equiv \frac{G_\infty}{2} \int_0^{t_0} [\gamma_{t_0^+} - \gamma_{t'}]^2 \frac{\partial \mathcal{G}(t - t')}{\partial t'} dt', \quad (7)$$

where we have ignored $\mathcal{G}(\cdot)t$ term in (6). Experimentally, the corresponding quantity has been measured using the calorimetry (Hasan and Boyce, 1993). They prepared glassy rubber samples with or without pre-stretching, and compared the exothermic heat upon slow warming. The pre-stretched sample, i.e. the sample containing the internal stress in the present context, released an excess heat. If this heat corresponds to the decrease of E_t^{int} in (7), the results implies that the mesoscale Hooke springs are *not* of entropic origin.

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